

Patent Application No. 10/524,455
Filing Date: February 10, 2005
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DECLARATION OF DR. ANDREAS SCHULTZ
Under 37 C.F.R. §1.132

Honorable Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

I, **ANDREAS SCHULTZ**, declare and state that:

1. I am a resident of the Federal Republic of Germany.
2. I am a citizen of the Federal Republic of Germany.
3. I am a chemist, having received a university degree in Chemistry from TU (Technical University) Braunschweig in Germany, in 2003.
4. Since 2008 I have been employed with SE Tylose GmbH & Co. KG ("SE Tylose"). My research for SE Tylose has generally focused on cellulose ethers. For the past 2 years my work has focused on cellulose ether product and process development.
5. I consider myself qualified by my knowledge of chemistry and cellulose chemistry and by my 6 / 2 years of experience in these technical fields.
6. I have personal knowledge of the above-captioned United States Patent Application 10/524,455.
7. I have read and understand the Official Action mailed on June 1, 2009 ("the Office Action").
8. I understand that Claims 1, 4, 6 through 9, 11, 12, 14 and 15 stand rejected over United States Patent No. 3,072,635 (US 635) to Menkart et al in light of United States Patent No. 4,366,070 (US 070) to Block.

9. To prove the beneficial properties imparted by the claimed process, I performed two experiments, referred to below as "Example 1" and "Comparative Example 2." Example 1 was conducted in accordance with the materials and methods of the claimed invention, in which cellulose ether is first admixed but not dissolved in either water or solvent and crosslinked with a compound having at least one aldehyde group and one acid group. In contrast, Comparative Example 2 was conducted in accordance with Example 1 of Block, except that glyoxylic acid (i.e. a compound having at least one aldehyde group and one acid group) was used in lieu of the glyoxal used in Block's Example 1.
10. As clearly illustrated by a comparison of the examples described below, the use of a dilute cellulose ether solution for crosslinking results in lump formation during subsequent cross-linked cellulose dissolution. Altogether unexpectedly, the more concentrated crosslinking methods of the claimed invention instead results in lump-free stirrability for the resulting cross-linked cellulose dissolution.
11. Example 1: In Example 1, 400 g of methylhydroxyethylcellulose (viscosity according to Hoeppler in 1.9% aqueous solution: 60,000 mPas) having a water content of 45% was mixed intensively for 30 minutes with 200 g of ice, 2.2 g of citric acid, 1.84 g of 50% aqueous sodium hydroxide solution and 7.5 g of 50% glyoxylic acid solution, comminuted using a disintegrator and milled wet. The milled product was dried for one hour at a temperature of 105 °C.
12. The crosslinked cellulose ether product of Example 1 was subsequently dissolved in water and had a solvation delay of 4.5 min, a final dissolving time to complete viscosity development of 30 minutes. The crosslinked ether product of Example 1 did not form lumps upon stirring into water.

13. Comparative Example 2: Comparative Example 2 was performed generally according to Example 1 from US 4366070 (at column 9, from line 15), except that glyoxylic acid was used as a crosslinker in lieu of the glyoxal in Example 1. An amount of 32.5 g of commercially obtained hydroxyethyl cellulose was initially dissolved in 1500 ml of water, resulting in a 2.1% cellulose solution.
14. Example 1 of US 070 indicates the formation of a 5 % cellulose solution; however, I found the upper limit of the cellulose concentration limited by viscosity level of the starting material. Specifically, I found that a solution with higher cellulose concentration was not stirrable. To the dissolved cellulose, 13.6 g of a 50% aqueous glyoxylic acid solution was added, and the pH was adjusted to 3.5 with diluted caustic soda.
15. The temperature was raised to 50 °C, and the solution was stirred for two hours. Example 1 of US 070 notes heating to 60 to 70 °C; however, I found that reaction with methylhydroxyethylcellulose has to be carried out at temperatures as low as possible because methylhydroxyethylcellulose precipitates at higher temperatures. The specification of US 070 notes that its crosslinking may be performed in the range of 50 ° to 100 °C at Column 5, lines 66 through 68, thus we remain within the teachings of US 070. A longer reaction time was then used to compensate for the lower temperature used in Comparative Example 2 in comparison to US 070 Example 1. After precipitation and removal of water with a total amount of 25 L of acetone, the product was air-dried, comminuted using a disintegrator and milled.
16. The crosslinked cellulose ether produced in Comparative Example 2 was subsequently dissolved in an aqueous solution and had a solvation delay of 1.5 min, a final dissolving time to complete viscosity development of 30 minutes. In contrast to Example 1, the crosslinked ether product of Comparative Example 2 formed lumps upon stirring into the aqueous solution.

17. Accordingly, a comparison of Example 1 and Comparative Example 2 demonstrate that the process of Block (i.e. US 070) provides cellulose with ethers with inferior properties. Comparative Example 2 was conducted in the way described in Example 1 of US 070, in which the cellulose ether to be crosslinked was completely dissolved in the water. In the inventive Example 1 the crosslinking is carried out in a heterogeneous system, in which the cellulose ether is not dissolved. In both Example 1 and Comparative Example 2 the same crosslinking agent was employed, namely glyoxylic acid. The properties of the resulting crosslinked cellulose ethers differed significantly, however.

18. The crosslinked cellulose ether according to Comparative Example 2 had a shorter solvation delay, and undesirable lumps were formed while stirring the crosslinked cellulose ether into water. In contrast, inventive Example 1 had a solvation delay which was three times longer (4.5 minutes versus 1.5 minutes) and could be stirred into water without formation of lumps. The improvement provided by the presently claimed process, in which cellulose ether is first admixed in water or solvent but not dissolved, was clearly unpredictable, yet highly advantageous.

Accordingly, I respectfully submit that Claims 1, 4, 6 through 9, 11, 12, 14 and 15 are patentable over US 635 and US 070, considered either alone or in combination.

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19. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statement may jeopardize the validity of the application or any patent issued thereon.

28 / 09 / 2009
Date

A. Schultz
Dr. Andreas Schultz